Population properties and carrier dynamics in a GaAs/(Al,Ga)As double-quantum-well superlattice investigated by time-resolved photoluminescence spectroscopy

L. Schrottke,[a] R. Hey, and H. T. Grahn
Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5-7, 10117 Berlin, Germany

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We have analyzed the electric-field-dependent subband population as well as the carrier dynamics in a double-quantum-well GaAs/(Al,Ga)As superlattice using time-resolved photoluminescence (PL) spectroscopy. Applying a rate equation model, the steady-state subband population of the majority carriers in the two quantum wells and the transfer coefficients for the minority carriers can be directly determined from measured time-dependent PL spectra. A comparison with results derived from steady-state PL investigations demonstrates that the dynamics of the minority carriers are essential in order to determine the population of the majority carriers. In the experiments, we used an *n–i–n* structure, in which electrons are the majority and holes are the minority carriers.
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For the investigation of light emitters on the basis of intersubband transitions, e.g., quantum cascade lasers (QCLs),[1] a quantitative evaluation of the electronic subband population is essential. Photoluminescence (PL) spectroscopy, which is a bipolar method as both the electron *and* hole concentrations determine the intensity, has been proven to be a powerful tool even for the investigation of such unipolar systems, if only one hole subband is involved in the interband transitions.[2–5] In more complex semiconductor heterostructures such as QCLs, so far the electron distribution has only been determined qualitatively through various optical methods.[6] However, for a precise quantitative discussion of the electric-field-dependent electronic subband population, the dynamics of electrons *and* holes has to be considered.

Previously, we have analyzed the population properties of a double-quantum-well (DQW) superlattice (SL), which served as a model system for QC structures, using continuous-wave PL spectroscopy.[7] One period of the SL consists of two quantum wells (QWs) and two barriers, which all have different thicknesses. While the variation of the barrier thicknesses results in different transfer rates, which are expected to lead to an occupation inhomogeneity between the electronic ground states of the adjacent wells, the difference in the well widths allows to identify the QWs following the thin or thick barrier (with respect to the direction of the electric field) through their PL energies. Using an appropriate set of rate equations, it was shown that, in order to meet the condition of weak excitation, the excitation intensity limit can easily be determined from the intensity dependence of the PL spectra. Assuming a homogeneous hole distribution, the electron population ratio was estimated from the PL intensity ratio as a function of the applied electric field. However, the assumption of a homogeneous hole distribution in DQW SLs is rather questionable for the following two reasons. First, the electric field leads to a redistribution of the photoexcited holes. Second, the hole subband for which the respective interband transition involves the electron subband with the larger population is depopulated faster.

In this letter, we investigate the dynamics of optically induced minority carriers in a DQW GaAs/(Al,Ga)As SL using time-resolved PL spectroscopy. It is shown that the subband population of the majority carriers, which is induced electrically by a steady-state injection current, can be directly determined from the time dependence of the PL intensities. Using a set of rate equations, the temporal behavior of the minority carriers, which are optically excited by ultrashort light pulses, is described. The majority carriers are considered to be independent of time. This assumption is valid as long as the optically excited density of the majority carriers is much lower than the electrically injected one, i.e., in the case of weak excitation. The experiments have been performed for a SL forming the intrinsic region of an *n–i–n* structure so that holes are the minority and electrons the majority carriers.

In the experiments, an $GaAs/Al_{0.3}Ga_{0.7}As$ SL with 20 periods was used. Each period consists of a 4 nm quantum well (QW_1), a 14 nm barrier, a 5 nm well (QW_2), and a 10 nm barrier. The photoexcitation was carried out by 200 fs pulses of a Ti:sapphire laser. The PL transients were detected using a streak camera with a nominal time resolution of 2 ps. Immediately after photoexcitation, the hole distribution can be assumed to be homogeneous so that the hole populations $p_1(0)$ and $p_2(0)$ in the corresponding QWs are equal. Therefore, the corresponding population ratio for the electrons can be directly determined from the ratio of the PL intensities. The excitation energy was tuned to 1.72 eV, which is below the energy gap of the barrier material, but well above the interband transition energies for both QWs so that homogeneous absorption can be assumed. For the investigated *n–i–n* structure, the temporal behavior of the PL spectra allows to determine the values of the electron population in each QW as well as the hole transfer rates through the barriers.

The rate equations for the hole sheet densities $p_1(t)$ and

[a]Electronic mail: lutz@pdi-berlin.de

$p_2(t)$ in QW_1 and QW_2, respectively, are written in the following form

$$\frac{dp_1(t)}{dt}=t_{21}p_2(t)-(r_1n_1+t_{12})p_1(t), \quad (1)$$

$$\frac{dp_2(t)}{dt}=t_{12}p_1(t)-(r_2n_2+t_{21})p_2(t). \quad (2)$$

The quantities t_{12} and t_{21} refer to the transfer coefficients per unit time of the holes between adjacent wells. n_1 and n_2 denote the electron sheet densities, while r_1 and r_2 refer to the respective recombination coefficients. Within this simple model, the spatial distribution of carriers is assumed to be laterally homogeneous and the same in each period of the SL. Therefore, we have to consider only a single period (QW_1 and QW_2). Since the electron concentration is solely determined by injection, the dynamics of the photoexcited electrons is neglected and assumed to be independent of time. However, the holes, which are only excited by the fs pulses, exhibit a temporal behavior.

The secular equation of the set of differential equations with constant coefficients in Eqs. (1) and (2) has two eigenvalues $\omega_{1,2}=-\alpha\pm\beta$, where

$$2\alpha=r_1n_1+t_{12}+r_2n_2+t_{21}, \quad (3)$$

$$2\beta=\sqrt{(2\Delta)^2+4t_{12}t_{21}}, \quad (4)$$

$$2\Delta=r_1n_1+t_{12}-(r_2n_2+t_{21}). \quad (5)$$

For homogeneous excitation so that the initial conditions are $p_1(0)=p_2(0)=p_0$, the analytic solution of Eqs. (1) and (2) is given by

$$p_1(t)=p_0e^{-\alpha t}\left[\cosh(\beta t)+\frac{t_{21}-\Delta}{\beta}\sinh(\beta t)\right], \quad (6)$$

$$p_2(t)=p_0e^{-\alpha t}\left[\cosh(\beta t)+\frac{t_{12}+\Delta}{\beta}\sinh(\beta t)\right]. \quad (7)$$

In order to determine the population of the electronic subbands in the two QWs, we consider the ratio $\rho(t)=I_1(t)/I_2(t)$ of the spectrally integrated PL time traces $I_i(t)$ of the two wells. At long times, it approaches a finite value rather than zero as the time traces do. The PL time traces $I_i(t)$ of QW_i are proportional to $r_in_i\,p_i(t)$ so that the ratio $\rho(t)$ can be written as

$$\rho(t)=\frac{r_1n_1p_1(t)}{r_2n_2p_2(t)}=\frac{r_1n_1}{r_2n_2}\,\frac{\beta+(t_{21}-\Delta)\tanh(\beta t)}{\beta+(t_{12}+\Delta)\tanh(\beta t)}. \quad (8)$$

In the limits of $t=0$ and $t\rightarrow\infty$, we obtain

$$\rho_0=\rho(0)=\frac{r_1n_1}{r_2n_2}, \quad (9)$$

$$\rho_\infty=\rho(\infty)=\rho_0\frac{\beta+t_{21}-\Delta}{\beta+t_{12}+\Delta}=-\rho_0\frac{\beta-t_{21}-\Delta}{\beta-t_{12}+\Delta}. \quad (10)$$

In Eq. (10), we used the definition (4) for β in order to arrive at the final expression. The experimental values of ρ_0 and ρ_∞ allow to determine the parameters r_1n_1, r_2n_2, t_{12}, and t_{21} provided that the eigenvalues $\omega_{1,2}$ are known. Solving Eqs. (9) and (10) for r_1n_1 and r_2n_2 using the expression for α and Δ as given in Eqs. (3) and (5), respectively, results in

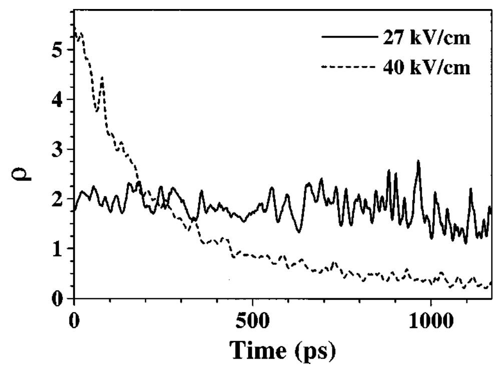

FIG. 1. Ratio $\rho(t)=I_1(t)/I_2(t)$ of the PL time traces of the narrower $[I_1(t)]$ and wider $[I_2(t)]$ QWs vs time for 27 and 40 kV/cm.

$$r_1n_1=-\omega_1\frac{\rho_\infty+\rho_0}{1+\rho_\infty}\quad\text{and}\quad r_2n_2=-\frac{\omega_1}{\rho_0}\,\frac{\rho_\infty+\rho_0}{1+\rho_\infty}. \quad (11)$$

In order to determine t_{12} and t_{21}, two linear equations have to be solved. The first equation is obtained by starting from Eq. (10) using $\Delta=\alpha-n_2r_2-t_{21}$ as well as the definition for $\omega_{1,2}=-\alpha\pm\beta$, while the second equation results from $\omega_1+\omega_2=-2\alpha$. Using Eq. (11) for r_1n_1 and r_2n_2, we obtain

$$t_{12}=\frac{\rho_0}{1+\rho_\infty}\omega_1-\frac{\rho_0}{\rho_0+\rho_\infty}\omega_2,$$
$$t_{21}=\frac{\rho_\infty}{\rho_0(1+\rho_\infty)}\omega_1-\frac{\rho_\infty}{\rho_0+\rho_\infty}\omega_2. \quad (12)$$

While the values ρ_0 and ρ_∞ can be directly obtained from experimental data, the determination of the eigenvalues $\omega_{1,2}$ requires a specific fitting procedure, for which, fortunately, neither the time scale nor the absolute values of the data need to be known. Our approach uses an auxiliary function $\theta(t,\omega)$, which is defined as

$$\theta(t,\omega)=\frac{\frac{d}{dt}[e^{-\omega t}I_1(t)]}{\frac{d}{dt}[e^{-\omega t}I_2(t)]}. \quad (13)$$

This auxiliary function has the property that $\theta(t,\omega=\omega_i)=\text{const.}$, i.e., time independent. This property is a consequence of the general representation for the solution $I_i(t)=A_{i,1}\exp(\omega_1 t)+A_{i,2}\exp(\omega_2 t)$ of the rate equation. Therefore, ω_i can be obtained by varying ω, until a time-independent auxiliary function is found.

Figure 1 shows the curves $\rho(t)$ for applied field strengths of $F=27$ and 40 kV/cm. The advantage of the approach is based on the fact that only the relative values ρ of the experimental data for $t=0$ and $t\rightarrow\infty$ have to be known. Although the curves $\rho(t)$ are less smooth than the time traces $I_i(t)$, the values for ρ_0 and ρ_∞ are expected to be more reliable than any fitting procedure for the time traces, since $I_i(t)$ contains four fitting parameters. In Fig. 2, the intensity ratio is shown as a function of electric field for different times (symbols). With increasing time, $\rho(F)$ decreases and its maximum shifts to lower electric fields. At long times, it approaches the value of ρ obtained in the stationary case (solid line), i.e., at continuous excitation.[7] The maximum value for $t=0$ is about three times as large as in the stationary

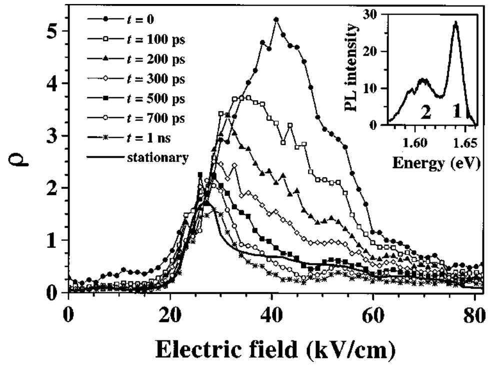

FIG. 2. $\rho = I_1/I_2$ vs applied electric field recorded at different times as indicated (symbols). For comparison, the field dependence of ρ for the stationary case is also shown (solid line). The inset displays a typical PL spectrum with two lines from the two QWs.

case. This result shows that the dynamics of the holes is essential even for a qualitative discussion of the subband occupation in such unipolar systems.

To obtain the electronic subband population and the hole transfer coefficients, $\omega_{1,2}$ are determined from the auxiliary function $\theta(t,\omega)$ by combining the calculation of the derivative with a smoothing procedure by convolution of the experimental curves with the derivative of the Gaussian curve. For the present sample, any electric-field dependence of the recombination coefficients r_1 and r_2 due to the quantum-confined Stark effect is negligible. Calculations using an envelope function approach in the effective-mass approximation show that for an electric field of 50 kV/cm the oscillator strength for both QWs is reduced by only 1%. Furthermore, due to the rather large thickness of the barriers, the coupling of the wave functions between the two wells at the heavy-hole resonances is very weak. Modifications of the oscillator strength would only occur within a very narrow range of the applied electric field of ± 1 V/cm. The field dependence of the population assuming $r_1 = r_2$ is shown in Fig. 3(a). While $n_2(F)$ (filled triangle) shows a moderate increase, $n_1(F)$ (clear square) clearly exhibits a maximum at about 45 kV/cm. The total population $n_1(F)+n_2(F)$ (filled circle) is, except for high electric fields, basically determined by n_1. Between 27 and 60 kV/cm, n_1 is significantly larger than n_2. For higher fields, both populations coincide. This is an indication for a transport mechanism, for which the thicknesses of the barriers do not play any role. We believe that the transport in this field range might be determined by carrier escape into continuum states and recapture.

The transfer coefficients for the holes shown in Fig. 3(b) are derived from Eq. (12). As expected, t_{12} (transfer through the thin barrier) is larger than t_{21} (transfer through the thick barrier). The field dependence of the transfer coefficients is much less pronounced than for n_1. In order to tentatively interpret the variation of t_{12} and t_{21}, we calculated the energetic positions of the hole subbands. At 4.6 kV/cm, the ground states in both QWs are resonant ($H1_nH1_w$). The resonances $H1_nH2_w$ ($H1_wH2_n$) of the lowest hole subband in the narrow (wide) well with the first excited hole subband in the wide (narrow) well occur at 38 kV/cm (50 kV/cm) via the thin (thick) barrier. The observed increase of t_{12} at 45 kV/cm agrees approximately with the resonance $H1_nH2_w$,

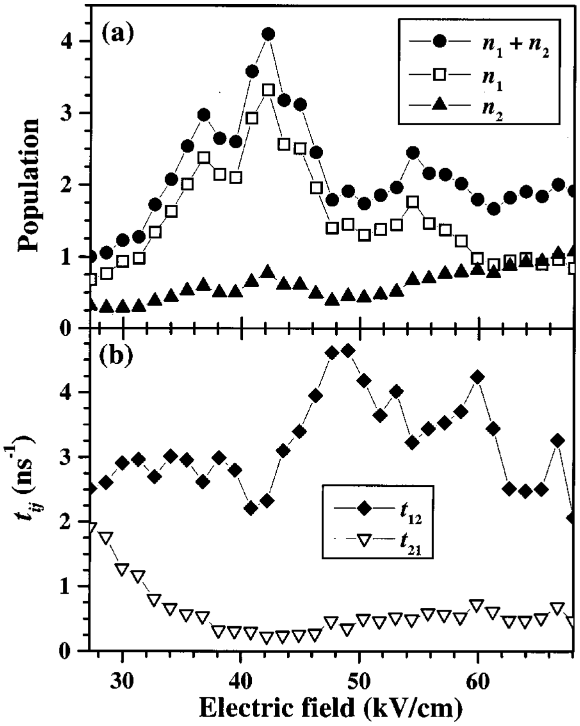

FIG. 3. (a) Electron population n_1 (□) and n_2 (▲) of the subbands of QW_1 and QW_2, respectively, and total population $n = n_1 + n_2$ (●) vs applied electric field. The values have been normalized to $n_1 + n_2$ at 27 kV/cm. (b) Hole transfer coefficients t_{12} ($QW_1 \rightarrow QW_2$, thin barrier) and t_{21} ($QW_2 \rightarrow QW_1$, thick barrier) vs applied electric field.

while the $H1_nH1_2$ resonance via the thick barrier (which does not occur for the thin barrier) at about 5 kV/cm may be responsible for the decrease of t_{21} below 40 kV/cm.

We have demonstrated that interband PL spectroscopy can be applied to determine the subband population of the majority carriers and the transfer coefficients of the minority carriers in more complex QW structures. The time dependence of the intensity ratio leads us to the conclusion that the dynamics of the minority carriers cannot be neglected. We observe significant deviations between the values estimated from steady-state PL spectroscopy and the ones derived from the time-resolved measurements. Further developments should also consider p–i–p structures, in which the role of minority and majority carriers is exchanged. Insofar as the intrinsic regions in a pair of n–i–n and p–i–p structures are identical, the electronic population ratio can also be determined from their transfer rates and compared with the directly determined values.

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